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### Abstract

Forward osmosis (FO) has emerged as a viable technology to alleviate the global water crisis. The greatest challenge facing the application of FO technology is the lack of an ideal draw solution with high water flux and low reverse salt flux. Hence, the objective of this study was to enhance FO by lowering reverse salt flux and maintaining high water flux; the method involved adding small concentrations of  $\text{Al}_2(\text{SO}_4)_3$  to a  $\text{MgCl}_2$  draw solution. Results showed that 0.5 M  $\text{MgCl}_2$  mixed with 0.05 M of  $\text{Al}_2(\text{SO}_4)_3$  at pH 6.5 achieved a lower reverse salt flux (0.53 gMH) than that of pure  $\text{MgCl}_2$  (1.55 gMH) using an FO cellulose triacetate nonwoven (CTA-NW) membrane. This was due possibly to the flocculation of aluminum hydroxide in the mixed draw solution that constricted membrane pores, resulting in reduced salt diffusion. Moreover, average water fluxes of 4.09 and 1.74  $\text{L/m}^2\text{-h}$  (LMH) were achieved over 180 min, respectively, when brackish water (5 g/L) and sea water (35 g/L) were used as feed solutions. Furthermore, three types of membrane distillation (MD) membranes were selected for draw solution recovery; of these, a polytetrafluoroethylene membrane with a pore size of 0.45  $\mu\text{m}$  proved to be the most effective in achieving a high salt rejection (99.90%) and high water flux (5.41 LMH) in a diluted draw solution.

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# **Exploration of an innovative draw solution for a forward osmosis-membrane distillation desalination process**

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## Abstract

Forward osmosis (FO) has emerged as a viable technology to alleviate the global water crisis. The greatest challenge facing the application of FO technology is the lack of an ideal draw solution with high water flux and low reverse salt flux. Hence, the objective of this study was to enhance FO by lowering reverse salt flux and maintaining high water flux; the method involved adding small concentrations of  $\text{Al}_2(\text{SO}_4)_3$  to a  $\text{MgCl}_2$  draw solution. Results showed that 0.5 M  $\text{MgCl}_2$  mixed with 0.05 M of  $\text{Al}_2(\text{SO}_4)_3$  at pH 6.5 achieved a lower reverse salt flux (0.53 gMH) than that of pure  $\text{MgCl}_2$  (1.55 gMH) using a FO CTA-NW membrane. This was due possibly to the flocculation of aluminum hydroxide in the mixed draw solution that constricted membrane pores, resulting in reduced salt diffusion. Moreover, average water fluxes of 4.09 and 1.74 LMH were achieved over 180 minutes, respectively, when brackish water (5 g/L) and sea water (35 g/L) were used as feed solutions. Furthermore, three types of MD membranes were selected for draw solution recovery; of these, a polytetrafluoroethylene membrane with a pore size of 0.45  $\mu\text{m}$  proved to be the most effective in achieving a high salt rejection (99.90%) and high water flux (5.41 LMH) in a diluted draw solution.

**Keywords:** Forward osmosis; draw solution; desalination; flocculation; membrane distillation; seawater

## 1. Introduction

Desalination has become a pragmatic approach to augment fresh water supplies in many coastal areas around the world (Khawaji et al. 2008). Large-scale desalination processes using thermal distillation (e.g. multi-stage flash and multi-effect distillation) or reverse osmosis (RO) have been widely applied to extract fresh water from brackish or seawater for fresh water provision (Schiermeier 2008, Semiat 2008). Thermal distillation desalination processes involve the phase change of water from liquid to vapor and vice versa to obtain desalted water; thus, they consume huge amounts of energy (i.e. mostly in the form of heating) to produce a volume of fresh water. In contrast, RO desalination exploits a high hydraulic pressure that pushes liquid water through a semipermeable membrane. The RO membrane is

permeable to water while retaining all particulates, virus, bacterium, and mostly dissolved salts (Elimelech & Phillip 2011). As a result, RO can produce fresh water directly from brackish or seawater with a significantly reduced energy consumption when compared to the thermal distillation processes. It is, however, noteworthy that RO desalination requires high-pressure pumps (i.e. hence costly duplex stainless steel tubing), and is highly susceptible to membrane fouling, thus involving considerable feed water pre-treatment and process maintenance.

Forward osmosis (FO) embodies notable attributes that render it a viable alternative to thermal distillation or RO for desalination applications. In FO, liquid water is extracted from a saline solution feed using a semipermeable membrane and a highly-concentrated draw solution (Achilli et al. 2010, Nguyen et al. 2016, Nguyen et al. 2013 ). Unlike in RO, the transport of fresh water through the membrane in FO is driven by the osmotic pressure difference between the feed and draw solutions. Consequently, FO desalination processes can be operated at a moderate hydraulic pressure, which obviates the need for high-pressure pumps and duplex stainless steel tubing as required by RO. More importantly, given the absence of a high hydraulic pressure, FO can directly filter saline feed solutions with less fouling propensity compared to RO. Membranes used in FO processes are also highly selective and therefore offer a high rejection of a wide range of contaminants as achieved by RO.

Draw solution plays a vital role in an FO desalination process (Achilli et al. 2010, Hau et al. 2014). During the FO process, salts from the draw solution reversely permeate through the membrane to the feed solution coincidentally with the transport of fresh water from the feed to the draw solution. The reverse salt flux results in a reduction in the osmotic pressure gradient across the FO membrane, thus reducing process water flux. The reverse diffusion of draw solutes to the feed solution also entails the subsequent replenishment of the draw solution to sustain the FO process water flux. Thus, an ideal draw solution is expected to offer

high water flux with a limited salt reverse diffusion. In addition, it is worth noting that FO must be coupled with another process for the regeneration of the draw solution and simultaneous extraction of fresh water. The draw solution regeneration process largely determines the energy consumption of the FO desalination process. Therefore, an ideal FO draw solution is also desired to be effectively regenerated with low energy requirements.

In recent decades, intensive studies have been conducted on FO draw solutions exploitation and their regeneration methods (Table S1). Notable examples include the study undertaken by McCutcheon et al. (2006). Using 1.6 M  $\text{NH}_4\text{HCO}_3$  as a draw solution, an FO process with a 29.25 g/L NaCl solution feed could achieve a high water flux ( $J_w$ ) of  $10.08 \text{ L.m}^{-2}.\text{h}^{-1}$  (McCutcheon et al. 2006). The diluted  $\text{NH}_3/\text{CO}_2$  draw solution could be effectively regenerated using a low-temperature distillation process and requiring relatively low energy consumption. However, a significant salt reverse flux (i.e.  $J_s = 18.20 \text{ gMH}$ ) was observed during the FO process when using 0.67 M  $\text{NH}_4\text{HCO}_3$  as a draw solution due to the small sizes of mono valent ions ( $\text{NH}_4^+$ ,  $\text{HCO}_3^-$ ) (Achilli et al. 2010). The reason was that the mono valent ions ( $\text{NH}_4^+$ ,  $\text{HCO}_3^-$ ) in draw solution was easy to reversely permeate through the FO membrane. To overcome this issue, Tan and Ng (2010) used divalent salts ( $\text{MgSO}_4$  and  $\text{CaCl}_2$ ) as the FO draw solutes, and reported a significantly lower salt reverse flux as compared to the process using  $\text{NH}_4\text{HCO}_3$ . The divalent salts draw solution was subsequently regenerated by an nano-filtration (NF) process. However, the NF process demonstrated a limited salts rejection at high  $\text{MgSO}_4$  and  $\text{CaCl}_2$  concentrations, thus inevitably leading to draw solution loss. Hau et al. (2014) employed ethylenediaminetetraacetic acid (EDTA) as a draw solution in an FO nanofiltration (FO-NF) hybrid system. The favorable solubility of EDTA in water helped the FO process achieve a noticeably high water flux (i.e. 12.60 LMH with a 0.7 M EDTA draw solution). Nevertheless, The FO process with EDTA draw solution experienced a high  $\text{Na}^+$  reverse flux, and the NF process could remain only 93% of EDTA during its regeneration. Similar to EDTA, polyelectrolytes of a series of polyacrylic acid

sodium salts (PAA-Na) were also explored given their high water solubility (Ge et al. 2012). Recently, several new draw solutes, including synthesized magnetic nanoparticles, hydrolyzed poly (isobutylene-*alt*-maleic acid), hexavalent phosphazene, and 2-methylimidazole-based organic compounds, have been tested for FO (Bai et al. 2011, Ge et al. 2014, Kumar et al. 2016, Ling et al. 2010, Lutchmiah et al. 2014, Stone et al. 2013 (a), Stone et al. 2013 (b), Yen et al. 2010). Although these draw solutes could be great potential in application, synthesizing the solutes is costly and recovery of the draw solutions is complex, consequently motivating the author to carry out this work. In this study, a novel draw solution for minimizing the reverse flux of ions during FO desalination to decrease the cost for replenishing lost draw solute.

This study aims to elucidate the feasibility of a brackish and seawater FO desalination process using a novel draw solution coupled with a membrane distillation (MD) process for its regeneration. The proposed novel draw solution was prepared by adding low  $\text{Al}_2(\text{SO}_4)_3$  concentration into  $\text{MgCl}_2$  solution. It was hypothesized that flocculated aluminum at optimal pH values in the draw solution could absorb ions and form an additional layer on the FO membrane surface, thus alleviating the reverse flux of the draw solutes to the seawater feed. Therefore, the influences of the draw solution pH and concentration on the FO process performance were first systematically evaluated using deionized (DI) water as the feed solution. Then, the FO desalination process with brackish water and seawater feeds using the novel draw solution was demonstrated. Finally, the regeneration of the proposed FO draw solution using an MD process was scrutinized. As a thermally driven membrane separation process, MD was envisaged to offer viable, low-cost draw solution regeneration to the FO process.

## **2. Materials and methods**

### **2.1. The draw and feed solutions**

The draw solution was prepared by dissolving mixtures of laboratory-grade  $\text{MgCl}_2:\text{Al}_2(\text{SO}_4)_3$  (Merck Co., Ltd., Germany) at molar ratios of 2:1, 6:1, 10:1, 14:1, and 20:1 in DI water at room temperature. The pH of the draw solution was then adjusted to 2.9 to 3.5, 5.0, 6.5, and 7.0 using NaOH. The prepared draw solutions were continually stirred for 24 hours prior to all FO experiments.

DI water and synthetic brackish water and seawater were used as feed solutions. DI water was used in the experiments to determine the optimal pH and concentration of the FO draw solutions. Synthetic brackish water and seawater were employed in the desalination demonstration of the FO process. The specifications of these feed solutions were provided in Table 1.



**Table 1. Properties of synthetic brackish water and seawater as feed solutions**

Feed solutions	Total dissolved solid, ppm	Viscosity, cp	Osmotic pressure, bar
Brackish water (5 g/ L NaCl)	5000	0.96	4.02
Seawater (35 g/ L NaCl)	35000	1.14	27.78

## 2.2. FO and MD membranes

Cellulose triacetate (CTA) nonwoven membrane (15×22 cm) obtained from Hydration Technology Innovations (HTIs OsMem™ CTA Membrane 121204, Albany, OR, USA) was utilized for the FO setup. The FO membrane had an active layer on the top of a support layer, and had a total thickness of 50  $\mu\text{m}$ . The FO membrane was relatively hydrophobic (Jin et al. 2012, Xie et al. 2012 (a)) with the determined contact angle of 60°–80°. In addition, at pH > 4.5, it was negatively charged (Xie et al. 2012 (b)).

Three types of polytetrafluoroethylene (PTFE) membranes from Ray-E Creative Co., Ltd. (Taiwan) were employed for the MD setup. Their pore sizes and contact angles are given in Table 2.

**Table 2. Pore size and contact angle of PTFE membranes used for membrane distillation.**

Membrane	Pore size ( $\mu\text{m}$ )	Contact angle ( $^{\circ}$ )
PTFE #1	0.1	126 $\pm$ 5
PTFE #2	0.45	114 $\pm$ 4
PTFE #3	1.0	126 $\pm$ 2

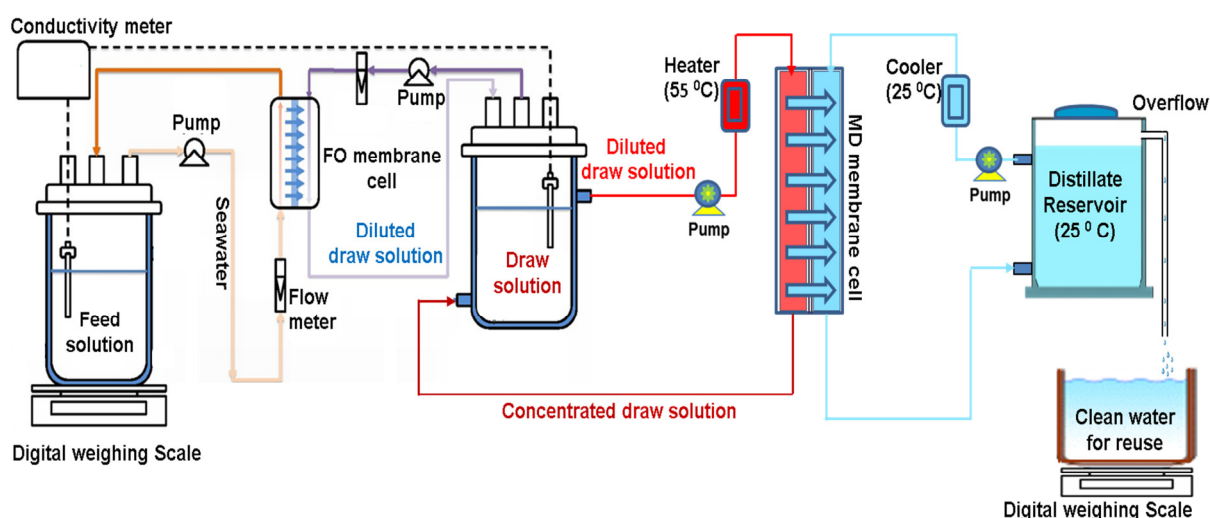
## 3.2. The lab-scale hybrid FO-MD system

The FO-MD hybrid system used in this study consisted of FO and MD membrane cells, FO feed and draw solution tanks, and a MD distillate reservoir (Figure 1). The FO membrane cell (FO Sterlitech) had two symmetric channels with width, length, and height of 4.5, 9.2, and 0.2 cm respectively, generating an effective membrane area of 41.40  $\text{cm}^2$  for mass transfer. Similarly, the MD membrane cell (Ray-E Creative Co., Ltd., Taiwan) was also

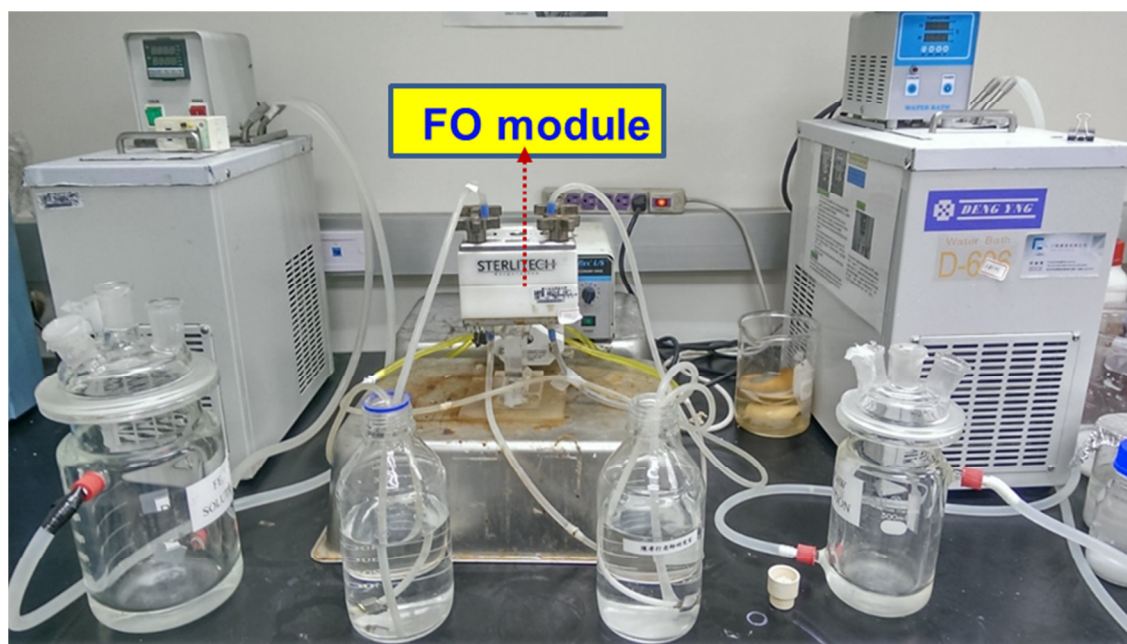
composed of two channels having width, length, and height, respectively, of 10.0, 10.0 and 0.3 cm. The active membrane surface for MD mass transfer was 100 cm<sup>2</sup>.

In this study, the FO and MD systems were operated separately to optimize desalination process. The feed solution (500 mL) and the draw solution (1000 mL) at room temperature (25 °C) were circulated through the FO cell under the same flow rate using two peristaltic pumps (Master Flux L/S Drive, Model 7518-00) (Figure 2). Conductivity and pH of the feed and the draw solution were regularly measured using sensors submerged in the solutions. The feed solution tank was placed on digital weighing scales (BW12KH, Shimadzu, Japan) connected to a computer for water flux measurements. Under the osmotic pressure difference across the FO membrane, water from the feed solution transferred through the membrane and diluted the draw solution.

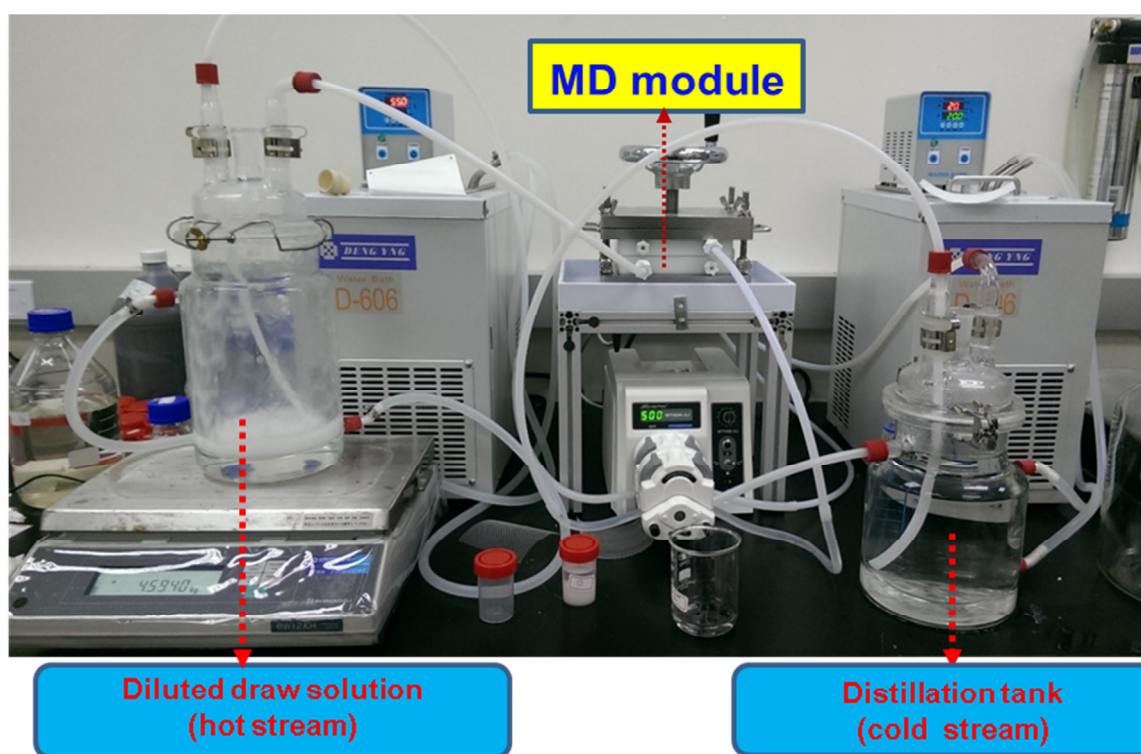
The diluted draw solution was then regenerated using the MD set up (Figure 3). During the MD regeneration, the diluted draw solution was heated to 55 °C and circulated through the MD feed channel. DI water (at 25 °C) was circulated on the other side of the membrane to condense water vapor permeated through the membrane from the diluted solution. The MD feed and distillate circulation rates were 0.083 m/s. The excess water from the distillate tank was regularly weighted for MD water flux measurement.



**Figure 1.** A schematic diagram of the lab-scale FO-MD hybrid system.



**Figure 2.** A photo of the FO setup for desalination of seawater.



**Figure 3.** A photo of the MD setup for water recovery.

### 2.3 Analytical methods

FO water flux ( $J_w$ ) was calculated as:

$$J_w = \frac{\Delta V}{A \Delta t} \quad (1)$$

where  $J_w$  was in L/m<sup>2</sup>-h (LMH),  $\Delta V$  was the feed volume change over a time interval  $\Delta t$  (hours), and  $A$  was the effective FO membrane area (m<sup>2</sup>). The reverse solute flux  $J_s$ , (in g/m<sup>2</sup>-h) of draw solution, was determined by the conversion of its electrical conductivity measured by a conductivity meter when MgCl<sub>2</sub> and Al<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> salt dissociated in its aqueous solution as follows:

$$J_s = \frac{V_t C_t - V_0 C_0}{A t} \quad (2)$$

where  $C_t$  and  $V_t$  were the concentration and volume of the feed solution, respectively, measured at time  $t$ , and  $C_0$  and  $V_0$  were the initial concentration and volume of the feed solution.

The specific reverse salt flux ( $J_s/J_w$  in g/L), which was the ratio of salt flux  $J_s$  and water flux  $J_w$ , was used to determine the amount of draw solute loss per a unit volume of produced water during an FO process.

The performance of the MD process for regeneration of the draw solution was evaluated using the process water flux and salt rejection. MD water flux was determined similarly to FO water flux, while MD salt rejection ( $R$ ) was calculated using Eq. (3):

$$R = (1 - \frac{EC_{distillate}}{EC_{feed}})100\% \quad (3)$$

where  $EC_{distillate}$  and  $EC_{feed}$  were the conductivity of the MD distillate and feed, respectively.

Viscosity and conductivity of the feed and draw solutions were determined using a Vibro Viscometer (AD Company, Japan) and conductivity meter (SensION156, Hach, China). The contact angles of the MD membranes were measured using the sessile drop method (i.e. droplet volume of 10±1 µL) on a CAM 100 (Opto-Mechatronics P Ltd., India). All measurements were conducted at room temperature (25 °C). The concentrations of Mg<sup>2+</sup>, Cl<sup>-</sup>, Al<sup>3+</sup>, and SO<sub>4</sub><sup>3-</sup> were analyzed using ion chromatography (Dionex ICS-90) and an ultraviolet-visible spectrophotometer (HACH Model DR-4000, Japan). The osmolality of

the solutions was measured using an osmometer (Model 3320, Advanced Instruments, Inc., USA), based on the freezing-point depression method (Gadelha et al. 2014). The draw solution's particle size was measured using a nanoparticle analyzer (SZ-100, Horiba, Japan).

### **3. Results and discussion**

#### **3.1. Effect of pH on water flux and reverse salt flux**

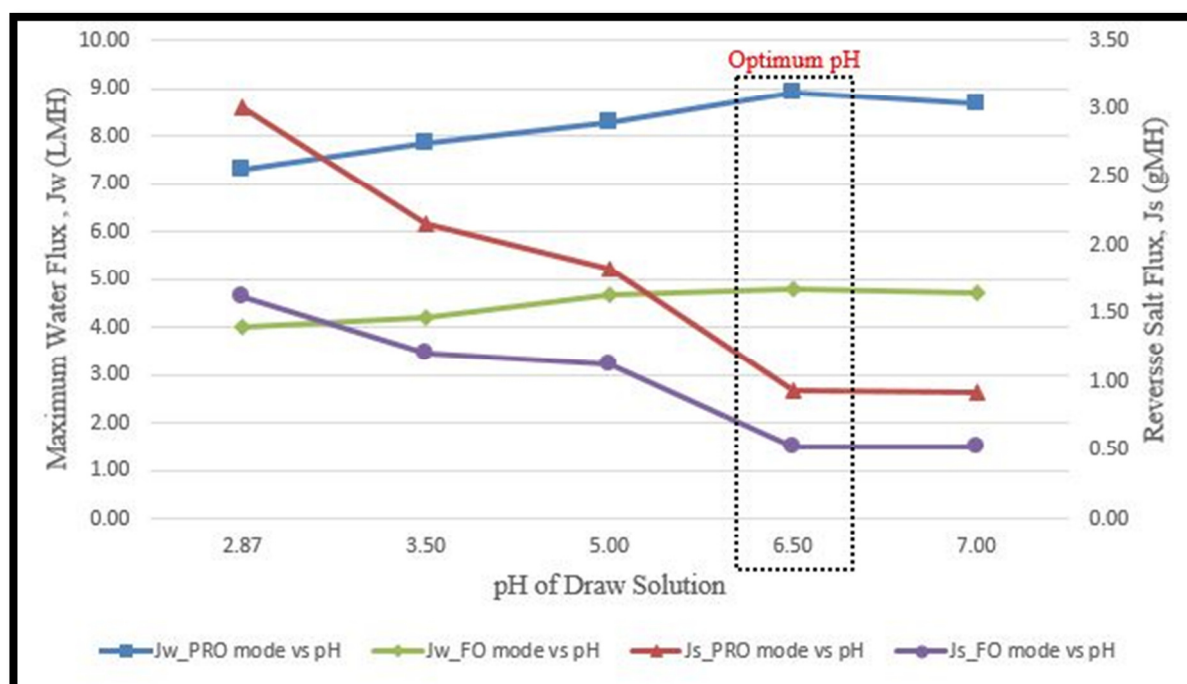
The values of water flux, reverse salt flux, specific reverse salt flux, and osmolality corresponding to different pH values of draw solution are shown in Figures 4 and 5. DI water was employed as a feed solution, 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$  was used as draw solution for the initial experiment. The initial pH of the given draw solution was found to be 2.87.

The water flux increased from 4.01 to 4.79 LMH in the FO mode (with the active layer facing the feed solution) and from 7.30 to 8.92 LMH in the PRO mode (with the active layer facing the draw solution) as the pH is increased from 2.87 to 6.50. This can be explained as follows: the addition of NaOH increased the osmolality of the draw solution. Also, the reverse salt flux decreased from 1.62 to 0.52 gMH in the FO mode and from 3.01 to 0.92 gMH in the PRO mode as the pH increased from 2.87 to 6.50.

This was due to a large amount of flocculation of aluminum hydroxide in the mixed draw solution at pH 6.50 (particle sizes of draw solution was large shown in Figure S1) that formed as a second layer on the FO membrane and constricted membrane pores, resulting in reduced salt diffusion (Figure S2). As the pH increased from 6.50 to 7.00, the water flux started to decrease in both FO and PRO modes, as shown in Figure 4. The decrease was due to excess formation of flocculation in the draw solution, which led to an increase in the solution's viscosity and thus to a decrease in the water flux. Therefore, the optimum pH condition for 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$  in both the FO and PRO modes was found

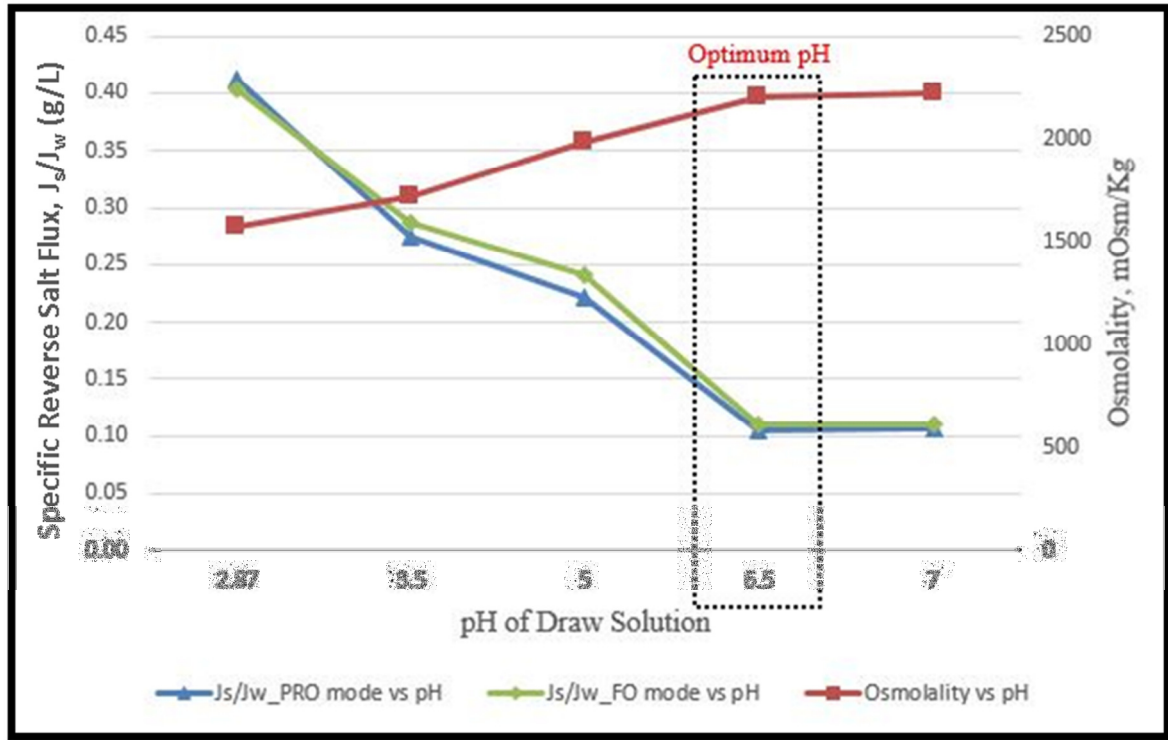
to be 6.5. The osmolality of the draw solution corresponding to the optimum pH was 2200 mOsm/kg.

At a pH of 6.5, the water flux and reverse salt flux of 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$  were 8.92 LMH and 0.94 gMH, respectively, in PRO mode. When only 0.5 M  $\text{MgCl}_2$  was used, the water flux and reverse salt flux were 8.27 LMH and 2.76 gMH, respectively, in PRO mode. Therefore when 0.05 M  $\text{Al}_2(\text{SO}_4)_3$  is added to the draw solution, the reverse salt flux decreases to a great extent due to the formation of flocculation, as discussed earlier.



**Figure 4.** Effect of pH on water flux and reverse salt flux. Feed solution: DI water; Draw solution: 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$ , flow rate: 0.5 L/min, experiment duration: 1 hr.



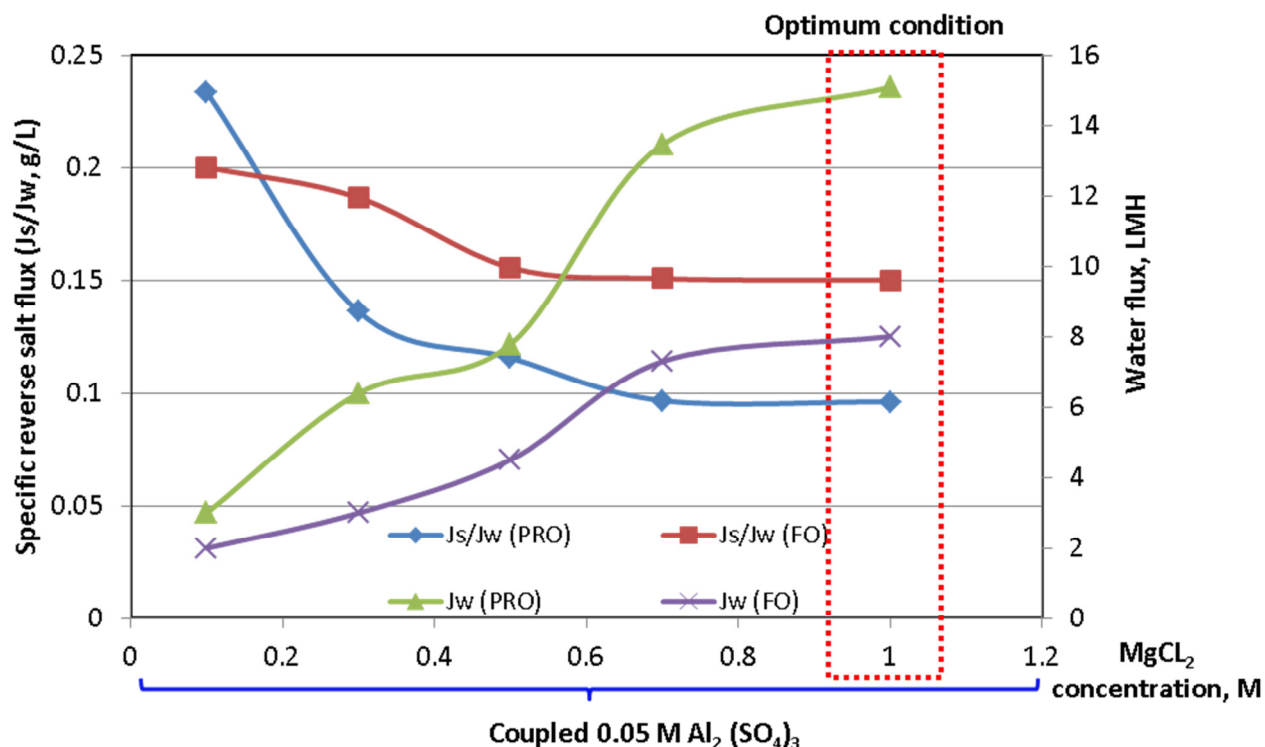


**Figure 5.** Effect of pH on specific reverse salt flux and Osmolality. Feed solution: DI water, Draw solution: 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$ , flow rate: 0.5 L/min, experimental duration: 1 hr.

### 3.2 Effect of draw solution concentration on water flux and reverse salt flux

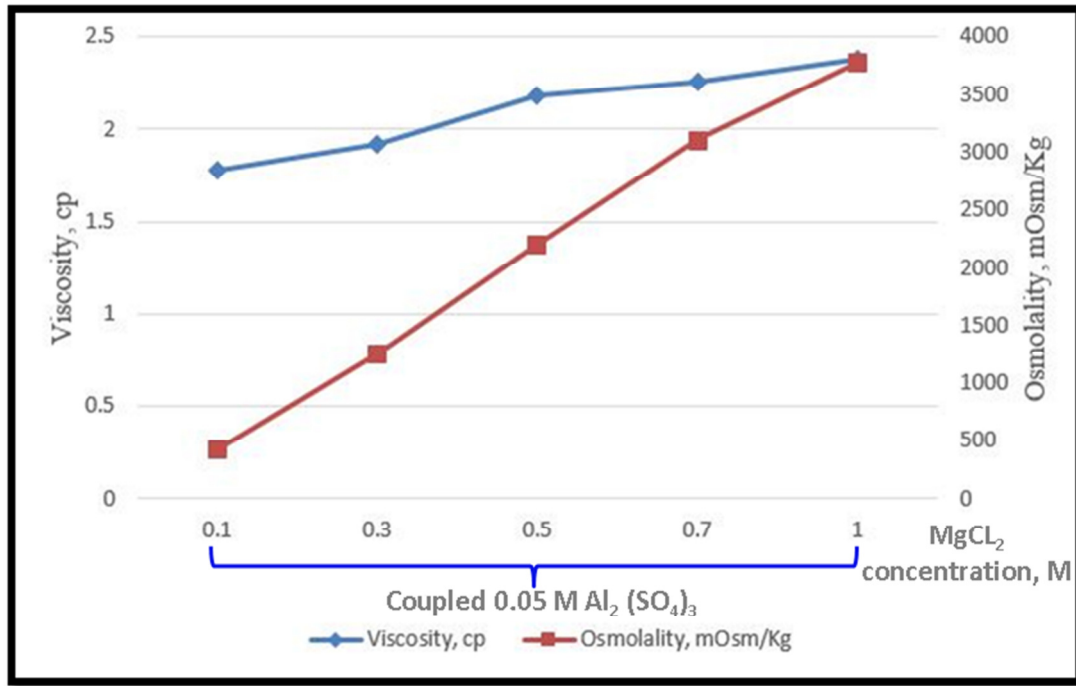
Figures 6 and 7 illustrate the reverse salt flux, water flux, viscosity, and osmolality for five draw solutions with various  $\text{MgCl}_2$  concentrations (from 0.1 to 1 M) coupled with a fixed  $\text{Al}_2(\text{SO}_4)_3$  concentration of 0.05 M. As shown in Figure 6, the water flux increased from 3.15 to 15.09 LMH in the PRO mode and from 1.71 to 8.18 LMH in the FO mode as the concentration of  $\text{MgCl}_2$  increased from 0.1 to 1 M. This can be explained by the sharp increase in the sample's osmolality. Also, the reverse salt flux increased from 0.52 to 2.65 gMH in the PRO mode and 0.299 to 1.61 gMH in the FO mode because of the increase in  $\text{Mg}^{2+}$  ions in the draw solution. Thus, the higher concentration of  $\text{MgCl}_2$  in the draw solution achieved a higher water flux; however, the nonlinearity of the variation of water flux with respect to the concentration was due to the effects of viscosity and internal concentration polarization (Figure 7).

Figure 6 shows that 1 M  $\text{MgCl}_2$  coupled with 0.05 M  $\text{Al}_2(\text{SO}_4)_3$  as draw solution achieved the lowest specific reverse salt flux ( $J_s/J_w = 0.096$  g/L in PRO and  $J_s/J_w = 0.151$  g/L in FO). This clearly demonstrated that a molar ratio of  $\text{MgCl}_2/\text{Al}_2(\text{SO}_4)_3 = 20$  was the optimal condition for the draw solution.



**Figure 6.** Effect of draw solution concentration on water flux and specific reverse salt flux. Feed solution: DI water; Draw solution: various  $\text{MgCl}_2$  concentrations from 0.1 to 1 M coupled with a fixed  $\text{Al}_2(\text{SO}_4)_3$  concentration of 0.05 M; flow rate: 0.5 L/min; experimental duration: 1 hr.





**Figure 7.** Effect of draw solution concentration on viscosity and osmolality.

### 3.3. Forward osmosis desalination process

To compare the efficiency levels of the desalination process with various feed solutions, three different feed solutions were used: DI water, brackish water (TDS = 5000 ppm), and sea water (TDS = 35000 ppm); 1 M MgCl<sub>2</sub> + 0.05 M Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> was used as a draw solution for desalination.

Figures S2 and S3 show the water flux decreased quickly in both FO and PRO modes when the osmotic pressure gradient between the draw and feed solutions decreased. During the first 30 min of the FO process, the DI water used as a feed solution achieved the highest water flux ( $J_w = 15.12 \text{ L/m}^2 \text{ h}$  in PRO mode and  $J_w = 8.09 \text{ L/m}^2 \text{ h}$  in FO mode), followed by brackish water with a TDS of 5000 ppm ( $J_w = 9.40 \text{ L/m}^2 \text{ h}$  in PRO mode and  $J_w = 5.03 \text{ L/m}^2 \text{ h}$  in FO mode), and sea water with TDS of 35 000 ppm ( $J_w = 3.95 \text{ L/m}^2 \text{ h}$  in PRO mode and  $J_w = 2.11 \text{ L/m}^2 \text{ h}$  in FO mode). When the desalination process was continued for additional time, the water flux was slightly decreased because of increases in the osmotic pressure of the feed solution (Table S2), which indicated that membrane fouling was neglected.

### 3.4 Recovery of diluted draw solution by membrane distillation

A membrane distillation process was tested to recover the diluted draw solution for reuse in the FO process. Three PTFE membranes with different pore sizes were used to determine the most suitable membrane. The water flux through these membranes is shown in Table 3. The results indicate that the highest water flux, 5.70 LMH, was achieved by the PTFE #3 of 1  $\mu\text{m}$  pore size. The water flux through PTFE #2 (pore size of 0.45  $\mu\text{m}$ ) and PTFE #1 (0 pore size of .1  $\mu\text{m}$ ) membranes were 5.41 and 4.95 LMH, respectively. The water flux increased with increases in pore size as the pore radius influenced the vapor transport; thus, a high pore radius tends to result in a high water flux. This confirmed the study conducted by Adnan et al. (2012), which discussed the influence of pore size on MD flux. The rejection percentage was found to be close to 100% in PTFE #1, PTFE #2(99.90%), and PTFE #3 membranes (99.06%). The reported result is consistent with previous studies by Duong et al. (2015), emphasizing the significant rejection of MD membrane due to partial vapor pressure differences across the membrane. As seen in Table 3, the difference in the water flux between PTFE#3 and PTFE#2 membranes was not appreciable; however, the PTFE#2 membrane retained a considerably higher amount of ions. Hence, the PTFE #2membrane was found to be most suitable for recovery of draw solution through membrane distillation with the conductivity rejection of approximately 100% and the concentration of conductivity in the permeate flux was as low as 93.8  $\mu\text{S}/\text{cm}$ , respectively, which was suitable for water reuse and drinking water.

**Table 3.** The effect of PTFE membrane pore sizes in MD system on removal efficiencies and water flux for diluted draw solution recovery.

MD Membrane	Conductivity in permeate ( $\mu\text{S}/\text{cm}$ )	Rejection (%)	MD Water flux $J_w$ , $\text{L}/\text{m}^2 \text{ h}$
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<b>PTFE #1</b>	69.2 ± 1.5	99.93	4.95 ± 0.11
<b>PTFE #2</b>	93.8 ± 1.9	99.90	5.41 ± 0.11
<b>PTFE #3</b>	893.0 ± 2.4	99.06	5.70 ± 0.12

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Diluted draw solution as initial feed with TDS= 61 483 mg/L, EC = 95 300  $\mu$ S/cm, pH = 6.5. Errors were based on the standard deviations of three replicate tests of three independent MD membranes.

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## Conclusions

A successful application of 1 M  $\text{MgCl}_2$  coupled with 0.05 M  $\text{Al}_2(\text{SO}_4)_3$  as a draw solution in a forward osmosis desalination process was demonstrated. The high solubility of the salt and flocculation created by  $\text{Al}_2(\text{SO}_4)_3$  not only provided a high osmotic pressure for high water flux, but also led to a reduced reverse salt flux as compared with numerous other inorganic salts. The chosen draw solution was able to desalinate brackish and sea water at water flux values of 4.09, and 1.74 LMH, respectively, using a CTA-NW membrane in FO mode. Furthermore, a PTFE #2 membrane (pore size of 0.45  $\mu\text{m}$ ) was selected as the most suitable membrane for recovering the diluted draw solutions with a solute rejection of approximately 100% and a MD water flux of 5.41 LMH.

## Acknowledgment

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## Figure Captions

**Figure 1.** A schematic diagram of the lab-scale FO-MD hybrid system.

**Figure 2.** A photo of the FO setup for desalination of seawater.

**Figure 3.** A photo of the MD setup for water recovery.

**Figure 4.** Effect of pH on water flux and reverse salt flux. Feed solution: DI water; Draw solution: 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$ , flow rate: 0.5 L/min, experiment duration: 1 hr.

**Figure 5.** Effect of pH on specific reverse salt flux and Osmolality. Feed solution: DI water, Draw solution: 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$ , flow rate: 0.5 L/min, experimental duration: 1 hr.

**Figure 6.** Effect of draw solution concentration on water flux and specific reverse salt flux. Feed solution: DI water; Draw solution: various  $\text{MgCl}_2$  concentrations from 0.1 to 1 M coupled with a fixed  $\text{Al}_2(\text{SO}_4)_3$  concentration of 0.05 M; flow rate: 0.5 L/min; experimental duration: 1 hr.

**Figure 7.** Effect of draw solution concentration on viscosity and osmolality.

## Tables

**Table 1**

Properties of synthetic brackish water and seawater as feed solutions

Feed solutions	Total dissolved solid, ppm	Viscosity, cp	Osmotic pressure, bar
Brackish water (5 g/ L NaCl)	5000	0.96	4.02
Seawater (35 g/ L NaCl)	35000	1.14	27.78

**Table 2**

Pore size and contact angle of PTFE membranes used for membrane distillation.

Membrane	Pore size ( $\mu\text{m}$ )	Contact angle ( $^{\circ}$ )
PTFE #1	0.1	$126 \pm 5$
PTFE #2	0.45	$114 \pm 4$
PTFE #3	1.0	$126 \pm 2$



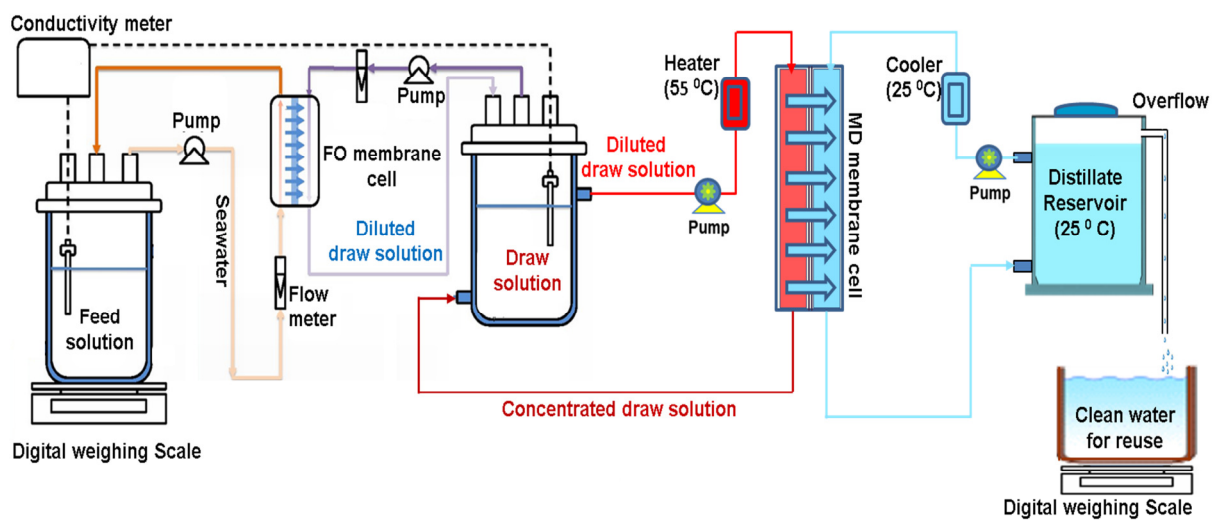
**Table 3**

The effect of PTFE membrane pore sizes in MD system on removal efficiencies and water flux for diluted draw solution recovery.

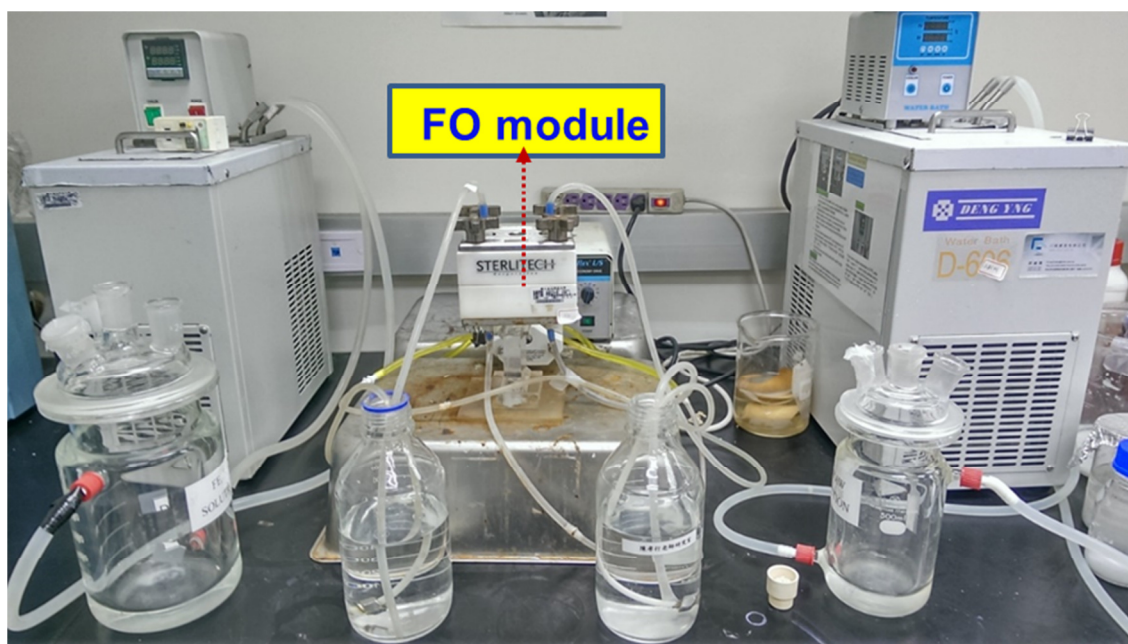
MD Membrane	Conductivity in permeate ( $\mu\text{S}/\text{cm}$ )	Rejection (%)	MD Water flux $J_w$ , $\text{L}/\text{m}^2 \text{ h}$
<b>PTFE #1</b>	$69.2 \pm 1.5$	99.93	$4.95 \pm 0.11$
<b>PTFE #2</b>	$93.8 \pm 1.9$	99.90	$5.41 \pm 0.11$
<b>PTFE #3</b>	$893.0 \pm 2.4$	99.06	$5.70 \pm 0.12$

Diluted draw solution as initial feed with TDS= 61 483 mg/L, EC = 95 300  $\mu\text{S}/\text{cm}$ , pH = 6.5. Errors were based on the standard deviations of three replicate tests of three independent MD membranes.

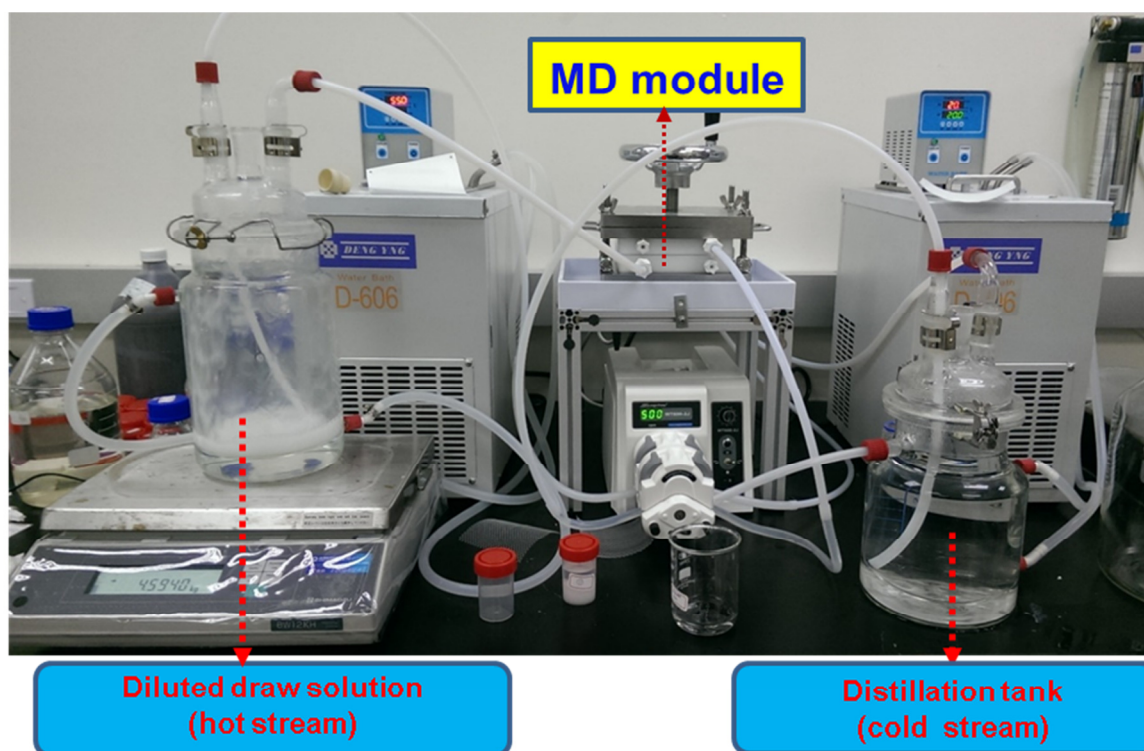
## Figures



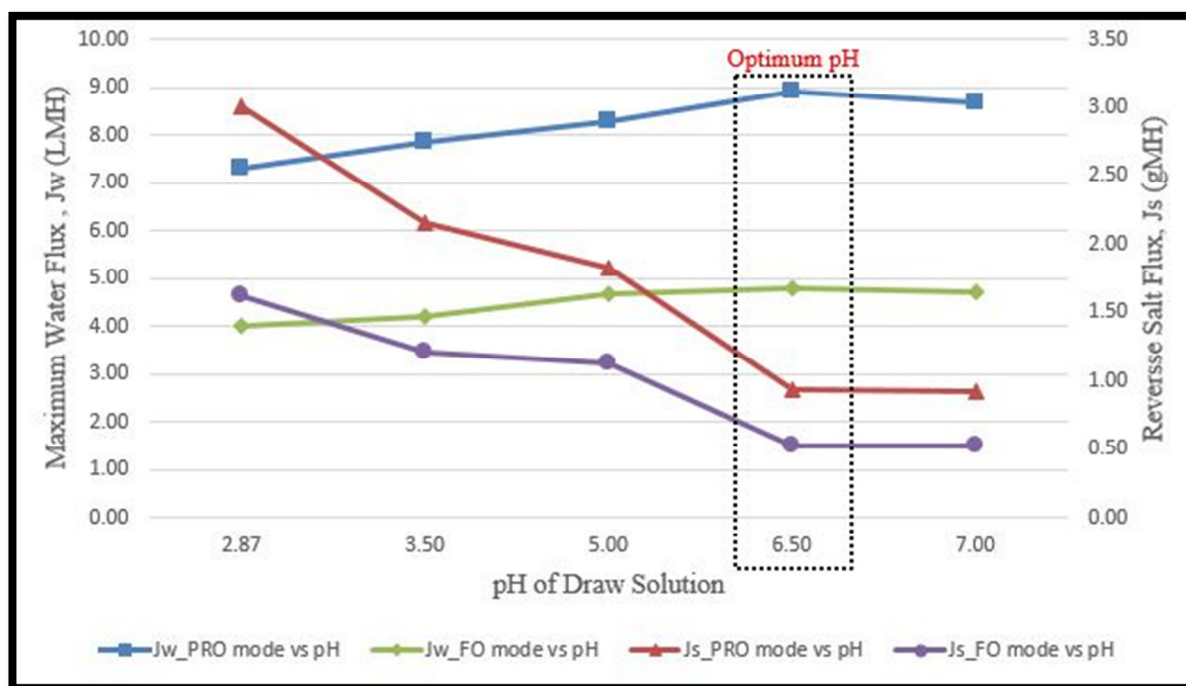
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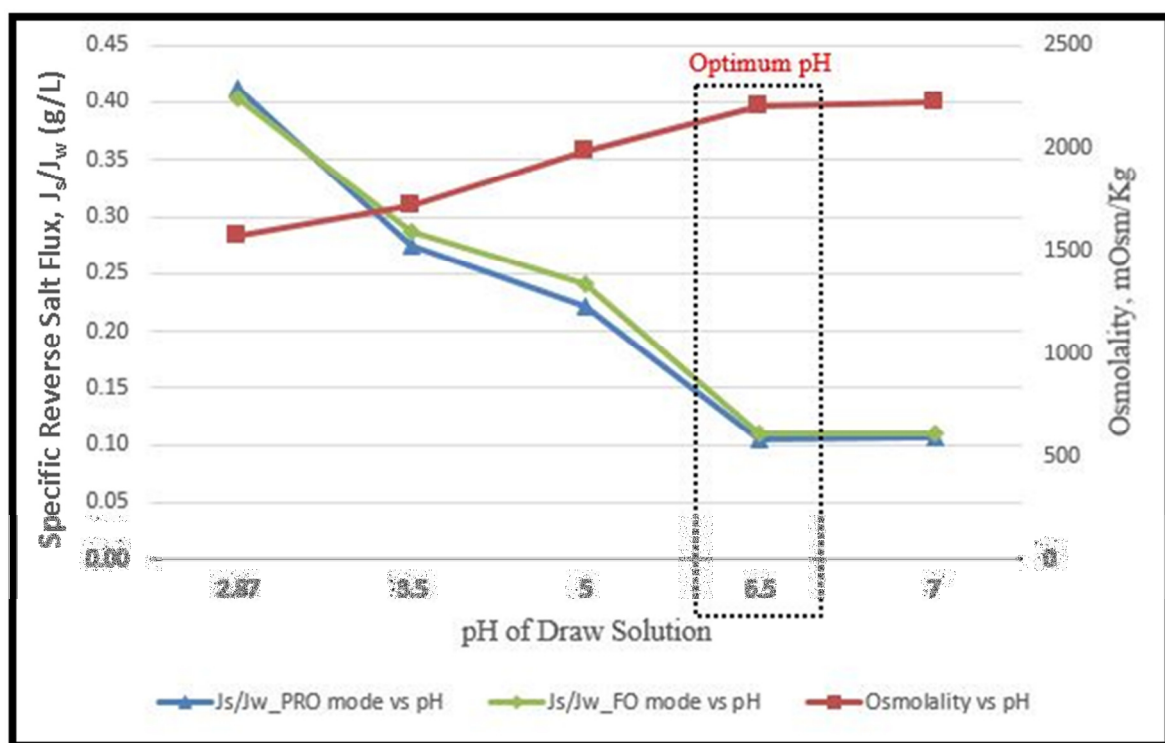
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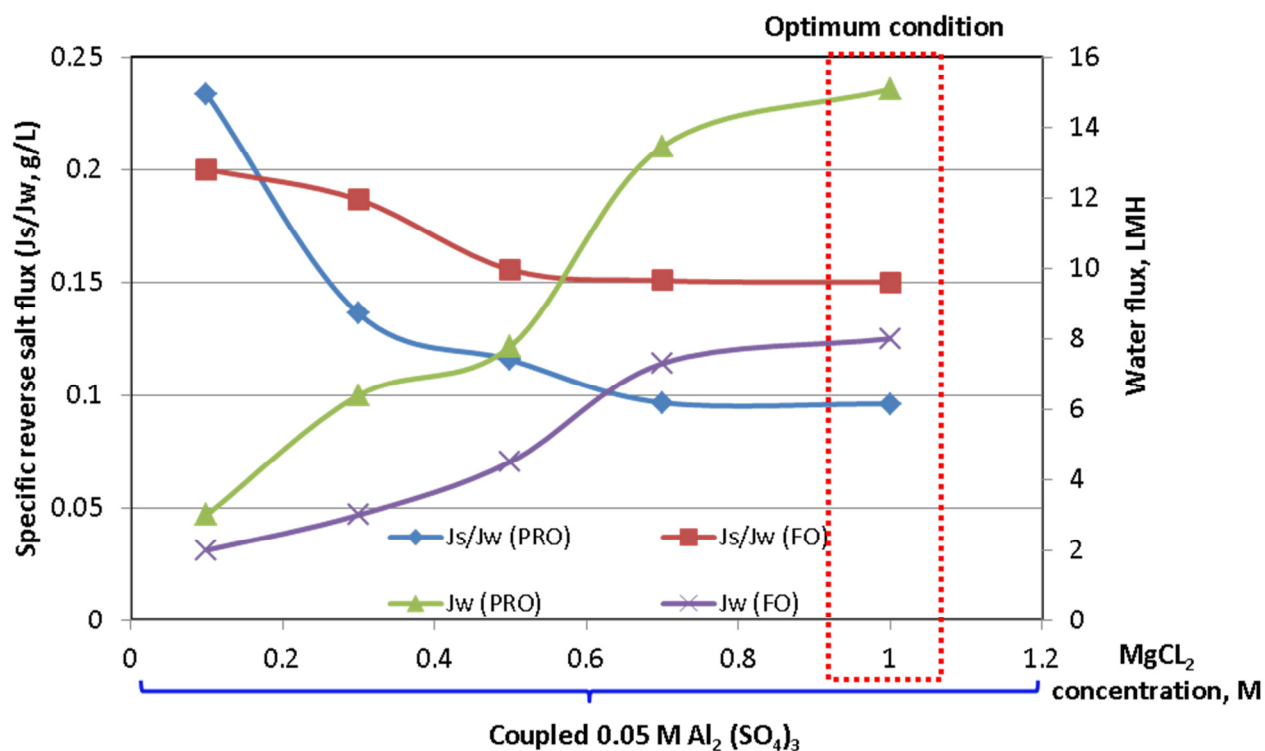
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**Figure 4.** Effect of pH on water flux and reverse salt flux. Feed solution: DI water; Draw solution: 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$ , flow rate: 0.5 L/min, experiment duration: 1 hr.

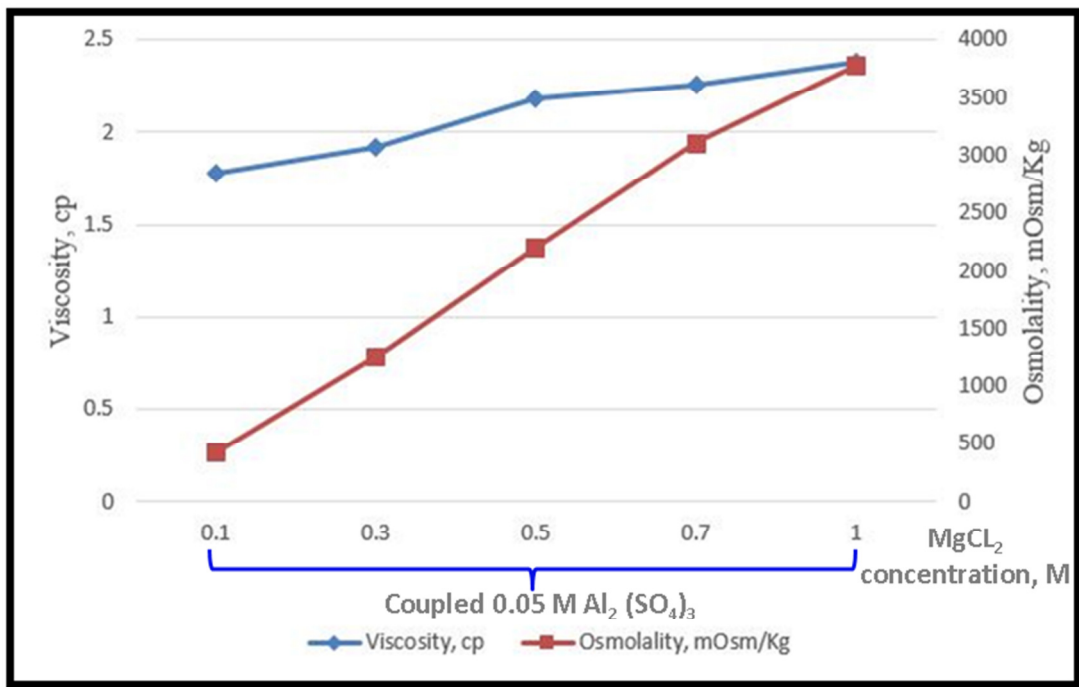


**Figure 5.** Effect of pH on specific reverse salt flux and Osmolality. Feed solution: DI water, Draw solution: 0.5 M  $\text{MgCl}_2$  + 0.05 M  $\text{Al}_2(\text{SO}_4)_3$ , flow rate: 0.5 L/min, experimental duration: 1 hr.



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Feed solution: DI water; Draw solution: various  $\text{MgCl}_2$  concentrations from 0.1 to 1 M coupled with a fixed  $\text{Al}_2(\text{SO}_4)_3$  concentration of 0.05 M; flow rate: 0.5 L/min; experimental duration: 1 hr.



**Figure 7.** Effect of draw solution concentration on viscosity and osmolality.



